

Europäisches Patentamt
European Patent Office
Office européen des brevets



(11)

EP 0 905 183 A1

(12)

EUROPEAN PATENT APPLICATION

(43) Date of publication:
31.03.1999 Bulletin 1999/13

(51) Int Cl.⁶: C08K 5/42

(21) Application number: 98307995.5

(22) Date of filing: 30.09.1998

(84) Designated Contracting States:
AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU
MC NL PT SE
Designated Extension States:
AL LT LV MK RO SI

(30) Priority: 30.09.1997 JP 282683/97

(71) Applicant: GENERAL ELECTRIC COMPANY
Schenectady, NY 12345 (US)

(72) Inventors:
• Saito, Akihiro
Utsunomiya City, Tochigi Prefecture 321 (JP)
• Yamaguchi, Shinichi
Utsunomiya City, Tochigi Pref. 321-0933 (JP)

(74) Representative: Szary, Anne Catherine, Dr. et al
London Patent Operation,
GE International, Inc.,
Essex House,
12-13 Essex Street
London WC2R 3AA (GB)

(54) Polycarbonate resin composition

(57) The present invention provides a polycarbonate (PC) resin composition which tends not to show amorphous-pattern contamination on the surfaces of molded articles, and is superior in terms of transparency, mechanical strength and weather resistance, etc. The PC resin composition contains (A) 100 parts by weight of a polycarbonate resin and (B) 0.0001 parts by weight or more, but less than 0.1 parts by weight, of a sulfonic acid phosphonium salt. Furthermore, the present invention also provides a PC resin composition

which additionally contains (C) 0.01 to 10 parts by weight of an ultraviolet absorbing agent.

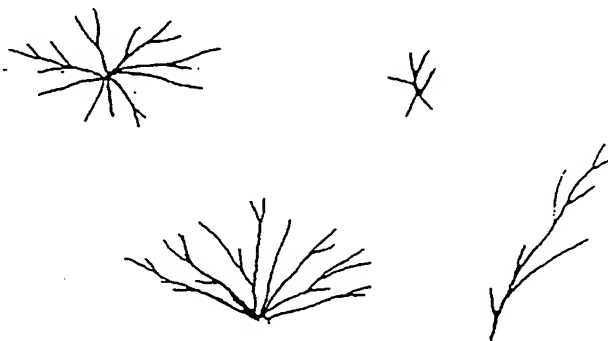
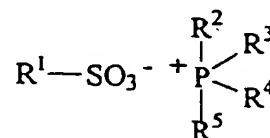


FIGURE 1

Description

[0001] The present invention concerns a polycarbonate (hereafter referred to as "PC") resin composition and a molded article consisting of the same.

[0002] PC resins are superior in terms of transparency, heat resistance and mechanical strength, and are therefore used in various fields. For example, PC resins are used in applications such as automobile head lamp lenses, covers and lenses for various illuminating devices, transparent films and transparent sheets, optical disks and optical disk cartridges, various parts used in office equipment and household appliances, and case materials used for storage or transportation, etc.

[0003] However, molded articles consisting of PC resins suffer from the problem of contamination with fine dust from the outside during use. Such contamination is not uniform overall; instead, areas where contaminants tend to adhere and areas where contaminants tend not to adhere form a non-uniform amorphous pattern, e. g., a fern leaf pattern or tree branch pattern as shown in Figure 1. As time passes, the contaminated areas become more conspicuous, thus damaging the commercial value of the molded article.

[0004] For example, Japanese Examined Patent Application No. Hei 7-39537 discloses a composition in which a sulfonic acid phosphonium salt and a phosphorous acid ester are mixed with a PC resin as a method of improving the anti-static properties of such a PC resin. Here, the sulfonic acid phosphonium salt is added at the rate of 0.1 to 20 parts by weight per 100 parts by weight of PC resin. In the abovementioned patent, it is considered that an anti-static effect cannot be obtained if the amount added is less than 0.1 parts by weight.

[0005] The object of the present invention is to provide a polycarbonate resin composition which tends to prevent contamination in an amorphous pattern on the surfaces of molded articles, and which is superior in terms of transparency, weather resistance and mechanical strength.

[0006] Figure 1 shows examples of shapes of amorphous-pattern contamination formed on the surfaces of molded articles.

[0007] The present inventors conducted diligent research concerning the prevention of contamination in molded articles consisting of PC resin compositions. As a result, the inventors made the following discovery: i.e., in cases where a sulfonic acid phosphonium salt is added to a PC resin, the weather resistance is lost if the amount added is too large. Accordingly, it was found that if an extremely small amount of a sulfonic acid phosphonium salt which would not conventionally be considered sufficient to provide an anti-static effect is added to a PC resin, not only is there no loss of transparency, weather resistance or mechanical strength, but surprisingly, the formation of contamination in an amorphous pattern is also suppressed. This discovery led to the perfection of the present invention.

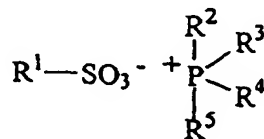
[0008] Specifically, the resin composition of the present invention is characterized by the fact that said compositions consists of (A) 100 parts by weight of a polycarbonate resin and (B) 0.0001 parts by weight or more, but less than 0.1 parts by weight, of a sulfonic acid phosphonium salt. The fact that contamination of PC resin molded articles in an amorphous pattern can thus be alleviated by the addition of an extremely small amount of a sulfonic acid phosphonium salt that would not be considered sufficient to provide an anti-static effect is something that could not have been predicted. Accordingly, it appears that the effect of sulfonic acid phosphonium salts in suppressing the formation of amorphous-pattern contamination in the present invention is based on a different mechanism from the conventional effect of providing anti-static properties.

[0009] The present invention also discloses a resin composition which includes (C) an ultraviolet absorbing agent in addition to the two components mentioned above. Conventionally, in the case of resin compositions containing (A) a PC resin and (C) an ultraviolet absorbing agent, the following problem has been encountered, i.e., when (B) a sulfonic acid phosphonium salt is added in order to prevent static electricity, the weather resistance drops conspicuously. However, it has been found that in cases where an extremely small amount of a sulfonic acid phosphonium salt is added as described above, amorphous-pattern contamination of molded articles can be alleviated without any loss of weather resistance.

[0010] In the present invention, aromatic polycarbonates manufactured by the universally known phosgene method or melt method, for example, see Japanese Unexamined Patent Application Disclosure No. Sho 63-215763 and Japanese Unexamined Patent Application Disclosure No. Hei 2-124934, can be used as the abovementioned (A) polycarbonate resin. Polycarbonate resins consist of a carbonate component and a diphenol component. Examples of precursor substances which can be used to introduce the carbonate component include phosgene and diphenyl carbonate, etc. Furthermore, examples of suitable diphenols include 2,2-bis(4-hydroxyphenyl)propane (so-called "bisphenol A" or "BPA"); 2,2-bis(3,5-dibromo-4-hydroxyphenyl)propane; 2,2-bis(3,5-dimethyl-4-hydroxyphenyl)propane; 1,1-bis(4-hydroxyphenyl)cyclohexane; 1,1-bis(3,5-dimethyl-4-hydroxyphenyl)cyclohexane; 1,1-bis(4-hydroxyphenyl)cyclododecane; 1,1-bis(3,5-dimethyl-4-hydroxyphenyl)cyclododecane; 4,4-dihydroxydiphenyl ether; 4,4-thiodiphenol; 4,4-dihydroxy-3,3-dichlorodiphenyl ether; and 4,4-dihydroxy-2,5-dihydroxydiphenyl ether, etc. These compounds may be used singly or in combination. In addition, compounds with three or more phenolic hydroxy groups may be used.

[0011] Alternatively, component (A) may be an aromatic copolyester carbonate. In addition to carbonate units originating in universally known aromatic diols, such compounds also have ester units originating in aromatic diols and aliphatic dicarboxylic acids with 6 to 18 carbon atoms. The phosgene method or melt method, which are universally known as methods of manufacturing aromatic polycarbonates, may be used to manufacture such compounds (see U.S. Patent No. 4,238,596, U.S. Patent No. 4,238,597 and U.S. Patent No. 3,169,121).

[0012] The abovementioned (B) sulfonic acid phosphonium salt used in the present invention is expressed, for example, by the following formula



In the above formula, R¹ indicates an alkyl group with 1 to 40 carbon atoms or an aryl group with 6 to 40 carbon atoms, and R², R³, R⁴ and R⁵ each independently indicate a group selected from a set consisting of hydrogen atoms, alkyl groups with 1 to 10 carbon atoms and aryl groups with 6 to 40, preferably 6 to 10 carbon atoms. Here the term alkyl group includes linear, branches and cyclic alkyl groups. Furthermore, the term aryl group includes alkylaryl and arylalkyl groups. Furthermore, such alkyl groups and aryl groups may be substituted by optional groups.

[0013] Examples of alkyl groups which may occur as R¹ include dodecyl groups, decyl groups, butyl groups and ethyl groups, etc. Examples of aryl groups include dodecylphenyl groups, phenyl groups, benzyl groups, phenethyl groups, tolyl groups and xylyl groups, etc. R¹ is preferably an aryl group.

[0014] Examples of alkyl groups which may occur as R² through R⁵ include methyl, ethyl, propyl and butyl groups, etc. Examples of aryl groups include phenyl groups, benzyl groups, phenethyl groups, tolyl groups and xylyl groups, etc.

[0015] Examples of desirable sulfonic acid phosphonium salts include tetraalkylphosphonium salts of dodecylsulfonic acid, and tetraalkylphosphonium salts of dodecylbenzenesulfonic acid, etc.

[0016] Component (B) is used in an amount which is at least 0.0001 parts by weight or more, preferably 0.0005 parts by weight or more, per 100 parts by weight of component (A) but less than 0.1 parts by weight, preferably less than 0.08 parts by weight, per 100 parts by weight of component (A). If the amount of component (B) that is used is too small, the effect of the invention cannot be obtained. On the other hand, if the amount used is too large, the weather resistance deteriorates; furthermore, discoloration occurs, the mechanical strength drops, and the external appearance of the molded article may be unsatisfactory.

[0017] In a resin composition containing the abovementioned components (A) and (B), contamination of molded articles in an amorphous pattern is suppressed while a superior transparency, weather resistance and mechanical strength are maintained.

[0018] The present invention also discloses a resin composition which contains (C) an ultraviolet absorbing agent in addition to the above mentioned components (A) and (B).

[0019] Any ultraviolet absorbing agent customarily used in PC resin compositions may be used as the above mentioned (C) ultraviolet absorbing agent. For example, benzotriazole type ultraviolet absorbing agents, benzophenone type ultraviolet absorbing agents or salicylate type ultraviolet absorbing agents, etc., may be used. Examples of benzotriazole type ultraviolet absorbing agents include 2-(2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-5'-t-butylphenyl)benzotriazole, 2-(2'-hydroxy-5'-t-octylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'-di-t-butylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'-di-amylobutylphenyl)benzotriazole, 2-(2'-hydroxy-3'-dodecyl-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'-dicumylphenyl)benzotriazole and 2,2'-methylenebis[4-(1,1,3,3-tetramethylbutyl)-6-(2H-benzotriazol-2-yl)phenol], etc. For example, a benzotriazole type ultraviolet absorbing agent is marketed by American Cyanamid Co. as UV5411. Furthermore, a benzophenone type ultraviolet absorbing agent is marketed by American Cyanamid Co. as UV531. Examples of salicylate type ultraviolet absorbing agents include phenyl salicylate, p-t-butylphenyl salicylate and p-octylphenyl salicylate, etc.

[0020] Component (C) is used in an amount which is at least 0.01 parts by weight or more, preferably 0.05 parts by weight or more, per 100 parts by weight of component (A) but less than 10 parts by weight or less, preferably 5 parts by weight or less, per 100 parts by weight of component (A). If the amount of component (C) that is used is too small, the light resistance deteriorates; on the other hand, if the amount used is too large, the heat resistance of the resin composition drops.

[0021] By combining the three components mentioned above, it is possible to obtain a resin composition in which contamination of molded articles in an amorphous pattern is suppressed while a superior transparency, weather resistance and mechanical strength are maintained.

[0022] Any of the resin compositions of the present invention may also contain phosphorus type stabilizers as optional

components in addition to the abovementioned components. For example, any of various stabilizers marked by various makers of stabilizers as oxidation inhibitors may be used as such phosphorus type stabilizers. Concrete examples of compounds which can be used include triphenyl phosphite, diphenylnonyl phosphite, tris(2,4-di-t-butylphenyl) phosphite, trisnonylphenyl phosphite, diphenylisooctyl phosphite, 2,2'-methylenebis(4,6-di-t-butylphenyl)octyl phosphite, diphenylisodecyl phosphite, diphenylmono(tridecyl) phosphite, 2,2'-ethylidenebis(4,6-di-t-butylphenyl) fluorophosphite, phenyldiisodecyl phosphite, phenyldi(tridecyl) phosphite, tris(2-ethylhexyl) phosphite, tris(isodecyl) phosphite, tris(tridecyl) phosphite, dibutyl hydrogenphosphite, triauryl trithiophosphite, tetrakis(2,4-di-t-butylphenyl)-4,4'-biphenylene diphosphite, 4,4'-isopropylidenediphenol alkyl (C₁₂ - C₁₅) phosphites, 4,4'-butylidenebis(3-methyl-6-t-butylphenyl)ditridecyl phosphite, bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite, bis(2,6-di-t-butyl-4-methylphenyl)pentaerythritol diphosphite, bis(nonylphenyl)pentaerythritol diphosphite, distearyl-pentaerythritol diphosphite, phenyl-bisphenol A pentaerythritol diphosphite, tetraphenyldipropylene glycol diphosphite, 1,1,3-tris(2-methyl-4-di-tridecylphosphite-5-t-butylphenyl)butane and 3,4,5,6-dibenzo-1,2-oxaphosphan-2-oxide, etc.

[0023] Available commercial products include Adekastab PEP-36, PEP-24, PEP-4C and PEP-8 (all trademarks, manufactured by Asahi Denka Kogyo K.K.), Irgafos 168 (trademark, manufactured by Ciba-Geigy Co.), Sandstab P-EPQ (trademark, manufactured by Sandoz Co.), Chelex L (trademark, manufactured by Sakai Kagaku Kogyo K.K.), 3P2S (trademark, manufactured by Ihara Chemical Kogyo K.K.), Mark 329K and Mark P (both trademarks, manufactured by Asahi Denka Kogyo K.K.) and Weston 618 (trademark, manufactured by Sanko Kagaku K.K.), etc.

[0024] It is desirable that phosphorus type stabilizers be added at the rate of 0.0001 to 1 parts by weight, preferably 0.001 to 0.5 parts by weight, per 100 parts by weight of PC resin.

[0025] Furthermore, hindered phenol type oxidation inhibitors, epoxy type stabilizers and sulfur type stabilizers, etc., may also be added. Examples of hindered phenol type oxidation inhibitors include n-octadecyl-3-(3',5'-di-t-butyl-4-hydroxyphenyl) propionate, 2,6-di-t-butyl-4-hydroxymethylphenol, 2,2'-methylenebis(4-methyl-6-t-butylphenol) and pentaerythritol-tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl) propionate, etc. Examples of epoxy type stabilizers include epoxidized soybean oil, epoxidized linseed oil, phenylglycidyl ether, allylglycidyl ether and 3,4-epoxycyclohexylmethyl-3',4'-epoxycyclohexane carboxylate, etc.

[0026] Furthermore, mold release agents may be added for the purpose of improving mold release characteristics. Examples of desirable mold release agents include silicon type mold release agents such as methylphenylsilicone oil, etc., and ester type mold release agents or olefin type mold release agents such as pentaerythritol tetrastearate, glycerol monostearate, montan acid wax and poly-alpha-olefins, etc.

[0027] Other customary additives may be added to all of the resin compositions of the present invention at the time of mixing or molding of the resin in amounts as necessary which do not have any deleterious effect on physical properties. For example, coloring agents (pigments or dyes), reinforcing agents (glass fibers, carbon fibers, etc.), fillers (carbon black, silica, titanium oxide, etc.), heat-resistant agents, oxidation inhibitors, weather-proofing agents, lubricants, mold release agents, plasticizer, flame retarding agents and fluidity enhancing agents, etc., may be added. Furthermore, dyes may be added in order to ameliorate yellowness in the blue direction.

[0028] There are no particular restrictions on the method used to manufacture the resin composition of the present invention; ordinary methods can be satisfactorily used. Generally, however, a melt mixing method is desirable. Small amounts of solvents may be used, but are generally not necessary. Examples of apparatus which can be used include especially extruders, Banbury mixers, rollers and kneaders, etc. Such machines may be operated in a batch operation or continuously. There are no particular restrictions on the order in which the components are mixed.

Examples

[0029] Below, the present invention will be described in greater detail in terms of working examples. However, the present invention is not limited to these examples.

[0030] Furthermore, the following substances were used in the working examples and comparative examples:

Component (A)

[0031] PC: polycarbonate, manufactured by Nippon GE Plastics K.K., trademark: LEXAN (intrinsic viscosity measured at 25°C in methylene chloride: 0.50 dl/g).

Component (B)

[0032] Sulfonic acid phosphonium salt: dodecylbenzene sulfonic acid tetrabutylphosphonium salt, manufactured by Takemoto Yushi K.K., trademark: EPA202.

Component (C)

[0033] Ultraviolet absorbing agent: benzotriazole type ultraviolet absorbing agent, 2-(2-hydroxy-5-t-octylphenyl)benzotriazole, manufactured by American Cyanamid Co., CYASORB UV5411.

Optional Components

[0034] Phosphorous acid ester: tris-(2,4-di-t-butylphenyl) phosphite, manufactured by Ciba-Geigy Co., Irgafos 168 (commercial name)

[0035] Hindered phenol type oxidation inhibitor: n-octadecyl-3(3',5'-di-t-butyl-4-hydroxyphenyl) propionate, manufactured by Asahi Denka Kogyo K.K., Adekastab AO-50.

[0036] Epoxy stabilizer: 3,4-epoxycyclohexylmethyl-3',4'-epoxycyclohexane carboxylate, manufactured by Daiseru Kagaku Kogyo K.K., Seroxide 2021P.

[0037] Furthermore, the various tests and evaluations performed in the examples and comparative examples were performed as follows

(1) Total Light Transmissivity

The total light transmissivity was measured according to ASTM D1003 for a 50 mm x 50 mm x 3 mm flat-plate injection molded sample

(2) Izod Impact Strength

The Izod impact strength was measured using a 1/8-inch notch according to ASTM D256.

(3) Surface Resistance

The surface resistance was measured according to ASTM D257 for a 50 mm x 50 mm x 3 mm flat-plate injection molded sample

(4) Weather Resistance Test

The discoloration (yellow index) after 1000 hours was measured using a fade-o-meter weather resistance tester (manufactured by Atlas Co.) for a 50 mm x 50 mm x 3 mm flat-plate injection-molded sample. Delta YI indicates the difference between the YI value after 1000 hours and the initial YI.

(5) Resistance to Discoloration

Following resistance for 10 minutes at a molding temperature of 280°C and a mold temperature of 80°C, a 50 mm x 50 mm x 3 mm flat plate was injection-molded, and the change in the yellow index was measured.

(6) Surface Contamination of Molded Article

A box form molded article (length 85 mm x width 200 mm x depth 15 mm, thickness 3 mm) which was injection-molded at a molding temperature of 280°C (set) and a mold temperature of 80°C (set) was allowed to stand indoors for 1 month (30 days), after which surface contamination was observed by visual inspection. In cases where the surface of the molded article showed contamination in an amorphous pattern (fern leaf pattern or tree branch pattern as shown in Figure 1), contamination was considered to be "present".

Examples 1 Through 4 and Comparative Examples 1 Through 4

[0038] In each case, the respective components were mixed in the proportions (weight ratio) shown in Table 1, and pellets were manufactured by extrusion using a single-shaft extruder (65 mm) set at 280°C, 100 rpm, 60 kg/hr (extrusion speed). Using the pellets thus obtained, injection molding was performed at a set temperature of 280°C and a mold temperature of 80°C. The molded articles thus obtained were subjected to various tests. The results are shown in Table 1

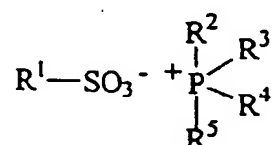
Table 1

	Working Example				Comparative Example			
	1	2	3	4	1	2	3	4
Components (parts by weight)								
P C	100	100	100	100	100	100	100	100
Sulfonic acid phosphonium salt	0.001	0.01	0.05	0.075	0.15	0.3	1	-
Ultraviolet absorbing agent	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Phosphorous acid ester	0.08	0.08	0.08	0.08	0.08	0.08	0.08	0.08
Hindered phenol	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Epoxy stabilizer	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Evaluations								
Total light transmissivity (%)	89	89	89	89	89	89	89	89
Izod impact strength (kg-cm/cm)	60	60	60	60	60	60	60	60
Surface resistances (ohms)	1×10^{15}	1×10^{15}	1×10^{15}	1×10^{15}	8×10^{14}	5×10^{14}	2×10^{12}	1×10^{15}
Delta YI (residence discoloration)	1.2	1.2	1.4	1.9	3.1	4.3	7	1.2
Delta YI (weather resistance test)	2.3	2.4	2.4	2.5	3.7	4.2	2.8	2.4
Surface Contamination	Absent	Absent	Absent	Absent	Absent	Absent	Absent	Present

The polycarbonate resin composition of the present invention tends not to show amorphous-pattern contamination on the surfaces of molded articles, and is superior in terms of transparency, mechanical strength and weather resistance, etc. Accordingly, the resin composition of the present invention is especially useful in applications such as automobile head lamp lenses, covers and lenses for various illuminating devices, transparent films and transparent sheets, optical disks and optical disk cartridges, various parts used in office equipment and household appliances, and case materials used for storage or transportation, etc.

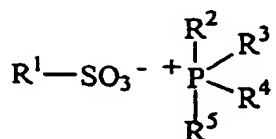
Claims

1. A polycarbonate resin comprising (A) 100 parts by weight of a polycarbonate resin and (B) from about 0.0001 parts by weight to about 0.1 parts by weight of a sulfonic acid phosphonium salt.
2. The resin of Claim 1, in which the amount of (B) is from to about 0.0005 to 0.08 parts by weight.
3. The resin of Claim 1 in which (B) is expressed by the following formula:



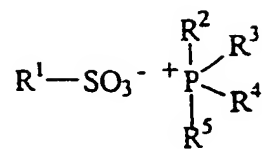
wherein R^1 represents an alkyl group with 1 to 40 carbon atoms or an aryl group with 6 to 40 carbon atoms, and R^2 , R^3 , R^4 and R^5 each independently represent a group selected from hydrogen atoms, alkyl groups with 1 to 10 carbon atoms and aryl groups with 6 to 40 carbon atoms.

4. The resin of Claim 1 which additionally comprises (C) from about 0.01 to about 10 parts by weight of an ultraviolet absorbing agent.
5. A molded article consisting of the resin of any preceding claim.
6. The molded article of Claim 5 which is an automobile head lamp lens.
7. The resin of Claim 1 in which (A) is a polycarbonate resin made by the melt process from bisphenol-A and diphenyl carbonate.
8. A polycarbonate resin consisting essentially of (A) 100 parts by weight of a polycarbonate resin and (B) from about 0.0005 parts by weight to about 0.08 parts by weight of a sulfonic acid phosphonium salt of the formula:



wherein R^1 represents an alkyl group with 1 to 40 carbon atoms or an aryl group with 6 to 40 carbon atoms, and R^2 , R^3 , R^4 and R^5 each independently represent a group selected from hydrogen atoms, alkyl groups with 1 to 10 carbon atoms and aryl groups with 6 to 40 carbon atoms.

9. A polycarbonate resin consisting essentially of (A) 100 parts by weight of a polycarbonate resin, (B) from about 0.0005 parts by weight to about 0.08 parts by weight of a sulfonic acid phosphonium salt of the formula:



wherein R^1 represents an alkyl group with 1 to 40 carbon atoms or an aryl group with 6 to 40 carbon atoms, and R^2 , R^3 , R^4 and R^5 each independently represent a group selected from hydrogen atoms, alkyl groups with 1 to 10 carbon atoms and aryl groups with 6 to 40 carbon atoms and (C) from about 0.01 to about 10 parts by weight of an ultraviolet absorbing agent.

10. The resin of claim 9 which additionally contains (D) a phosphate type stabilizer in the amount of from about 0.0001 to about 1 part by weight per 100 parts by weight of (A).

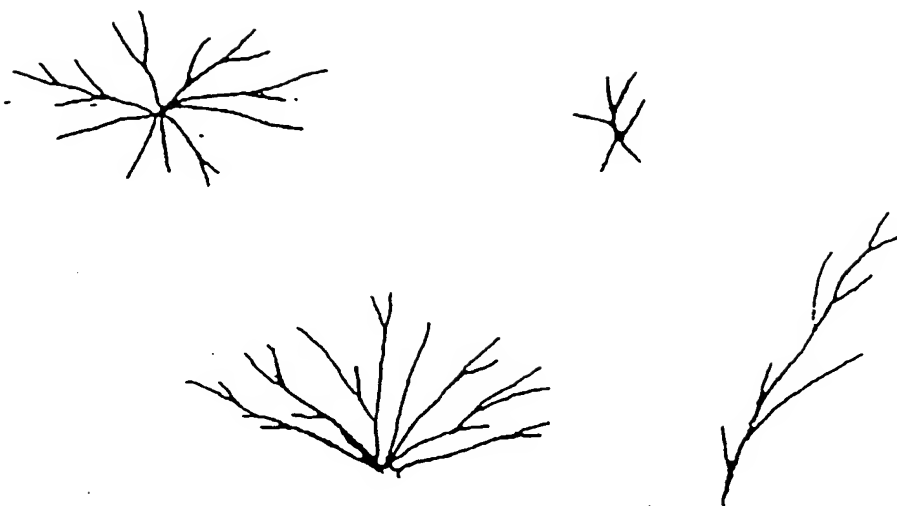


FIGURE 1



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 98 30 7995

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	EP 0 640 646 A (TEIJIN LTD) 1 March 1995 * page 15, line 58 - page 16, line 1 * * claims 1-4 *	1-10	C08K5/42
X	US 4 093 589 A (FACTOR ARNOLD ET AL) 6 June 1978 * column 7, line 42 - line 46 * * claims 1-4 *	1-10	
X	DATABASE WPI Section Ch, Week 9332 Derwent Publications Ltd., London, GB; Class A23, AN 93-252931 XP002083861 & JP 05 171024 A (MITSUBISHI KASEI CORP) , 9 July 1993 * abstract *	1-10	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C08K
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 14 December 1998	Examiner Siemens, T
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document			

EPO FORM 1503 (3.12.94) (P4/C01)

**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 98 30 7995

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

14-12-1998

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0640646 A	01-03-1995	JP 7062077 A	07-03-1995
		JP 7126374 A	16-05-1995
		JP 7165905 A	27-06-1995
		US 5486555 A	23-01-1996
		US 5494952 A	27-02-1996
		US 5668202 A	16-09-1997
US 4093589 A	06-06-1978	AU 504057 B	27-09-1979
		AU 2993377 A	26-04-1979
		BR 7800761 A	28-08-1979
		DD 132350 A	20-09-1978
		DE 2746906 A	10-08-1978
		FR 2379573 A	01-09-1978
		GB 1579079 A	12-11-1980
		JP 53097050 A	24-08-1978
		NL 7711472 A	07-08-1978
		SU 886751 A	30-11-1981

EPO FORM P0489

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

THIS PAGE BLANK (USPTO)